

**REMOVAL OF SULPHATE IN WASTE  
RECYCLE AGGREGATE USING *Bacillus subtilis***

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by

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## LIST OF ABBREVIATIONS

|                  |  |
|------------------|--|
| ANOVA            | Analysis of Variance                             |
| ASR              | Alkali silicate reaction                         |
| BOD <sub>5</sub> | Biochemical oxygen demand (five days)            |
| C-S-H            | Calcium – Silicate - Hydrate                     |
| CDW              | Construction and demolition waste                |
| CFU              | Colony forming unit                              |
| CIDB             | Construction Industry Development Board          |
| COD              | Chemical oxygen demand                           |
| DEF              | Delayed ettringite formation                     |
| DO               | Dissolved Oxygen                                 |
| EEF              | Early ettringite formation                       |
| EPS              | Extracellular polymeric substance                |
| FTIR             | Fourier-transform infrared                       |
| HDPE             | High density polyethylene                        |
| IBS              | Industrialised Building System                   |
| IC               | Ion Chromatography                               |
| ICP-OES          | Ion Coupled Plasma-Optical Emission spectrometry |
| INWQS            | Interim National River Water Quality Standard    |
| ITZ              | Interfacial transition zone                      |
| OD               | Optical Density                                  |
| ODD              | Oven dry density                                 |
| PZC              | Point Zero Charge                                |
| RCM              | Recycled cement mortar                           |

|      |                                  |
|------|----------------------------------|
| SDD  | Surface dry density              |
| SEM  | Scanning Electron Microscope     |
| TDS  | Total Dissolved Solid            |
| TSS  | Total suspended solid            |
| TWCA | Treated waste concrete aggregate |
| WCA  | Waste concrete aggregate         |
| XRD  | X-ray Diffractometric            |
| XRF  | X-ray Florescence                |



# **PENGURANGAN SULFAT DALAM SISA KITAR SEMULA AGREGAT MENGUNAKAN *Bacillus subtilis***

## **ABSTRAK**

Sisa konkrit agregat (WCA) berpotensi tinggi untuk diguna semula sebagai bahan kitar-semula kerana ia boleh digunakan untuk pelbagai fungsi. Lekatan simen tampal pada permukaan WCA menghadkan keupayaan WCA untuk dikitar-semula sebagai sebahagian daripada bahan asas pembangunan. Faktor limitasi ini ialah rendah ketumpatan tetapi tinggi penyerapan air serta kandungan sulfat. Kandungan sulfat di dalam pembentukan tundaan ettringit (DEF) pada lekatan simen tampal yang tinggi penyerapan air menjurus kepada pengembangan DEF dan membentuk retakan mikro menyebabkan kehilangan jisim dan memberi kesan kepada kekuatan mekanikal (kekuatan mampatan) konkrit. Objektif utama kajian ini adalah untuk merawat WCA dengan menggunakan *Bacillus subtilis* sebelum dikitar-semula sebagai simen mortar yang dikitar-semula (RCM) yang baru. Ciri-ciri WCA dikenalpasti terlebih dahulu sebelum ujian perlekatan *B. subtilis*. Keberkesanan *B. subtilis* untuk merawat WCA dinilai berdasarkan empat faktor pembolehubah iaitu kepekatan *B. subtilis*, nisbah *B. subtilis*: WCA, potensi zeta dan pengekal masa. WCA terawat (TWCA) daripada rawatan WCA oleh *B. subtilis* digunakan semula sebagai sebahagian daripada bahan asas pembuatan RCM. Kekuatan mampatan RCM seterusnya dinilai untuk menentukan kualitinya. Keputusan menunjukkan WCA mempunyai ketumpatan yang rendah, penyerapan air yang tinggi, terkarbonat dan kaya ettringit. WCA mengandungi keterlarutan sulfat sebanyak  $0.53 \pm 0.06\%$  dan  $47.76 \pm 8.43\%$  kandungan lekatan simen tampal. Kelakuan pelunturan elemen-elemen bukan organik dalam WCA menunjukkan Ca dan  $\text{SO}_4^{2-}$  sebagai element utama dengan kepekatan masing-masing

antara 10.06 mg L<sup>-1</sup> sehingga 149.15 mg L<sup>-1</sup> dan 30.78 mg L<sup>-1</sup> sehingga 38.90 mg L<sup>-1</sup>. Kandungan sulfat dalam WCA telah berjaya dikurangkan kepada 81.5% oleh *B. subtilis* pada kepekatan *B. subtilis* 0.2 x10<sup>8</sup> CFU mL<sup>-1</sup>, nisbah *B. subtilis* dan WCA 1:1, pengekalan masa meningkat sehingga 4d<sup>-1</sup> dan tanpa perubahan pada potensi zeta *B. subtilis* atau WCA. Pembinaan RCM menggunakan RCA mampu menggantikan agregat konkrit semula-jadi kerana mempunyai kekuatan mampatan yang lebih tinggi (26MPa) berbanding simen motar yang biasa (20MPa). Kajian ini menunjukkan lekatan sel *B. subtilis* pada WCA mampu mengurangkan kandungan sulfat dalam WCA dengan keluasan permukaan lebih besar untuk mengelak pembentukan rekahan mikro pada RCM.

## REMOVAL OF SULPHATE IN WASTE RECYCLE AGGREGATE USING

*Bacillus subtilis*

### ABSTRACT

Waste concrete aggregate (WCA) is highly potential to be re-utilised as a recycled material as it is applicable to numerous functions. Attached cement paste on WCA surface has limited the potential WCA to be recycled as a part of raw construction materials. Low density, high in water absorption and sulphate content are identified as the major contributing factors for this limitation. The sulphate content in delayed ettringite formation (DEF) from high water absorption of attached cement paste leads to DEF expansion and forms a micro-crack, causing a mass loss and affects the mechanical strength (compressive strength) of the concrete. The main objective of the present study was to treat WCA by incorporating the use of *Bacillus subtilis* before it was reused in recycled cement mortar (RCM) production. The characteristics of WCA were previously identified prior to the *B. subtilis* adhesion test. The efficiency of *B. subtilis* to treat WCA was evaluated based on four variable factors i.e. *B. subtilis* concentration, *B. subtilis*: WCA ratio, zeta potential, and retention time. Treated WCA (TWCA) from the WCA treatment by *B. subtilis* was reused as a part of RCM production. The compressive strength of RCM was subsequently evaluated to determine its quality. Results revealed that WCA had a low density, high water absorption, carbonated by attached cement paste and rich in ettringite. WCA contained  $0.53 \pm 0.06$  % of soluble sulphate and  $47.76 \pm 8.43$  % of attached cement paste. Leaching behaviour of inorganic element demonstrated Ca and  $\text{SO}_4^{2-}$  as major elements, ranging from  $10.06 \text{ mg L}^{-1}$  to  $149.15 \text{ mg L}^{-1}$  and  $30.78 \text{ mg L}^{-1}$  to  $38.90 \text{ mg L}^{-1}$ , respectively. The sulphate content in WCA was successfully reduced to 81.5% by

*B. subtilis* when *B. subtilis* concentration was at  $0.2 \times 10^8$  CFU mL<sup>-1</sup>, the *B. subtilis*: WCA ratios was 1:1 and the retention time was increased up to 4 d<sup>-1</sup> without changing the zeta potential of *B. subtilis* or WCA. The construction of RCM using TWCA has a potential to replace the natural concrete aggregate due to its higher compressive strength (26 MPa) compared to normal cement mortar (20 MPa). The study indicated that *B. subtilis* efficiently reduced sulphate in a bigger surface area of WCA to avoid the surface micro-crack formation on RCM.

## CHAPTER 1

### INTRODUCTION

#### 1.1 Background

Construction and demolition waste (CDW) is defined as a waste arises from construction, renovation and demolition activities which comprising land excavation and formation, civil and building construction, site clearance, roadwork and building renovation (Shen et al., 2004; Poon & Chan, 2007; Tam et al., 2008). In Malaysia, high amount of waste generated from CDW has become a serious problem due to the rapid development in the construction industry to cope the demand of housing, major infrastructure project and commercial developments (Nasaruddin et al., 2008; Siti & Noor, 2008; Begum et al., 2010).

The common disposal practice for CDW in Malaysia is illegal dumping at landfills, roadside and tropical mangrove swamp (Yahaya & Narsen, 2008; Murali, 2011; Nagapan et al., 2012; Tan, 2012). This practice has caused a land shortage and negatively impact the tourism industry (Pascual & Cladera, 2004). Furthermore, a large amount of CDW at the landfill has resulted in an emission of hydrogen sulphide ( $H_2S$ ), the colourless gas produced when vicinity microbes reacted with non-segregated CDW. As a consequence, it creates an odour problem, further causing an adverse effect to the human health (Jang & Townsend, 2001; O'Connell, 2005). Although the  $H_2S$  gas is potentially to be diluted by the surrounding ambient air, the released gas is considerably large to create the odour problem (Jang & Townsend, 2001). Nowadays, an awareness concerning the negative impact from CDW has risen among local communities in Malaysia (Begum et al., 2006). Moreover, the production

of construction materials from natural resources has been increasingly encouraged by policy makers for sustainability purpose (Oikonomou, 2005; Pappu et al., 2007). Therefore, European commission has revealed that CDW needs to be minimised and well-managed since such waste has been categorised as one of the main wastes at landfill and potentially risky to the environment (European Commision Report, 2002).

Reduction of CDW and conservation of natural resources by recycling are among the main targets in achieving the sustainable manner in construction industry (Nagapan et al., 2012). In order to achieve these, European communities have provided a proper guideline towards environmental protection and waste management policy particularly for construction industry (European Commisison Report, 2002). In Malaysia, Construction Industry Development Board (CIDB) has implemented Industrialised Building System (IBS) towards the sustainable development with zero waste concept (Kamarul & Zuhairi, 2007). One of the aims is to promote recycling practices by re-manufacturing and reusing CDW materials, of which the raw aggregate is replaced to a newly recycled product (Kamarul & Zuhairi, 2007).

There are many types of physical waste categorised as CDW e.g. concrete, ceramic, metal, wood, brick and block, tile, timber, asphalt, gypsum, paper, glass, soil and sand (Jand & Townsend et al., 2001; Faridah et al., 2004; Bianchini et al., 2005; Oikonomou, 2005; Vegas et al., 2011; Lachimpadi et al., 2012; Lee et al., 2013). Numerous studies have demonstrated the replacement of raw aggregates with CDW in producing different types of new recycled products (concrete, cement mortar and pavement block) with a great mechanical strength (Evangelista & de-Brito, 2007). Waste concrete is a major solid waste generated from construction industry worldwide

(Faridah et al., 2004; Oikonomou, 2005; Kamarul & Zuhairi, 2007). This physical waste has completely damaged and is crushed into waste concrete aggregate (WCA) potentially used in cement mortar production (Nagapan et al., 2012). The production of recycled products from CDW requires a mixture of raw coarse aggregate, cement and WCA of which the WCA is used to replace the sand as recycled concrete aggregate (Evangelista & de-Brito, 2007). However, utilisation of WCA without any pre-treatment process tends to produce a low quality of a cement mortar material due to the several drawbacks of WCA. Previous studies claimed that WCA has low density, high in water absorption as well as sulphate content compared to raw concrete aggregate and virgin soil due to the presence of attached cement paste (Evangelista & de-Brito, 2007).

Low density of WCA affects the compressive strength of recycled cement mortar, while, high water absorption and sulphate content provides a conducive environment for Delayed Ettringite Formation (DEF) which initiates the development of micro-crack (Collepari, 2003; Isgor & Razaqpur, 2005; Tam et al., 2007). These drawback characteristics are not only affected the compressive strength, but causes deterioration of recycled cement mortar particularly for long term used (Tam et al., 2007; Padmini et al., 2009).

Various initiatives have been done by researchers on WCA either by chemical or physical treatments in order to overcome such drawbacks. Admixture of superplasticiser in cement mortar mix has enhanced the compressive strength of recycled cement mortar, but corrosion with embedded steel reinforcement has caused this chemical treatment inappropriate to be applied (Binici, 2007). Another chemical

treatment was dissolution of attached cement paste onto WCA surface by a solution of hydrochloric acid (de Juan & Gutierrez, 2009). Although this treatment aimed to get rid a source drawback of the WCA (attached cement paste), but utilisation of acid has indirectly degraded composition of aggregate (Nagataki et al., 2000).

Similar results were seen in the physical treatment. The addition of fly ash and blast furnace slag in concrete and cement mortar mix were used to overcome the low density of WCA which beneficial for compressive strength. However, this treatment was not suitable for other drawbacks. These materials consist of pozzolana component that possessed cementitious properties for enhancement on cement hydration. However, these additives had increased the retardation rate of cement hydration due to high water demand which subsequently delayed the hardening process (Cheah & Ramli, 2011). Furthermore, high sulphate content in fly ash with the range of 2.20% to 12.50% was not suitable to be used together with WCA because its high possibility to initiate the DEF (Naik et al., 2003). The replacement of raw concrete aggregate with WCA at an appropriate ratio is another physical treatment to overcome the low density of WCA by enhancing the compressive strength of recycled product (Batayneh et al., 2007; Evangelista & de-Brito, 2007).

These physical and chemical treatments have improved on the mechanical strength of recycled product due to the low density of WCA but both treatments did not reduce the water absorption and sulphate content. Yet, none of the research focusing on reducing the sulphate content and water absorption in WCA by *Bacillus subtilis*.



A biological treatment by utilising *B. subtilis* in a field of concrete technology have proven that this genus able to survive in high alkaline environment ( $\text{pH} \geq 12$ ) of concrete. It could heal a micro-crack up to 2.7 cm in depth of the concrete surface and increased 40% compressive strength of the recycled product (Van Tittelboom et al., 2010; Gupta et al., 2013). Previous research claimed that *B. subtilis* was used to fill in the micro-crack on the concrete surface by calcium carbonate precipitation (Mohannadoss et al., 2015). Sulphate in WCA as well as an essential anions are vital in biochemical reaction and metabolic activities of *B. subtilis* (Ghosh et al., 2005). Therefore, the up-take of sulphate in WCA by *B. subtilis* as a pre-treatment prior to WCA utilisation as a cement mortar material could avoid the development of internal DEF and micro-crack, which simultaneously improve the quality of WCA.

## **1.2 Problem Statement**

Despite its high potential in the production of new recycled products, several drawbacks of WCA have been recognized such as low density, high water absorption as well as high sulphate content (Evangelista & de-Brito, 2007). These characteristics are often become the limiting factors for WCA to be recycled which influenced by the attached cement paste on the surface of WCA (Juan & Gutierrez, 2009; Vegas et al., 2011; de-Brito & Nabajyoti, 2013).

Sulphate content is one of the major problems in producing recycled products (cement mortar and concrete) because its presence could deteriorate the concrete (Collepari, 2002). Briefly, the presence of sulphate in the concrete may cause a phenomenon known as DEF, a needle-like structure of crystal, causing the micro-crack, mass loss and reducing the material strength (Irrassar, 2002; Hu et al., 2006).

All the above mentioned problems are due to the formation of weak interfacial transition zone (ITZ) between the cement and the aggregate particle caused by DEF (Tam et al., 2007). In addition, oxidation of pyrite (cement component) also results in the release of sulphate and subsequently enhances the DEF (Lee et al., 2005). Moreover, continuous exposure of concrete to water or under humid air is sufficient to support DEF (Collepardi, 2003). Ambient humidity of concrete, exposure of concrete under the sun and rain are also deteriorating following the micro-crack problem (Famy et al., 2001; Graf, 2007).

The use of bacteria such as *B. subtilis* has been introduced and investigated by many researchers to improve the quality of the recycled products. Such bacteria are able to improve the compressive strength and fill the void spaces by precipitation of calcium carbonate ( $\text{CaCO}_3$ ) (Sahoo et al., 2016). Application of *B. subtilis* on WCA is able to reduce the maximum sulphate content from DEF since *B. subtilis* is highly adaptable to high alkalinity of concrete and could consume the sulphate for its biochemical reaction and metabolic activities (Ghosh et al., 2005). By far, to the best of our knowledge, no work has been done on the importance of biotic and abiotic factors on WCA such as zeta potential, bacteria concentration and the availability of inorganic elements (Bhaskar, 2016). Impurity of sulphate and high water absorption in concrete structures causes a micro-crack formation that shorten the service life.

Therefore, utilisation of *B. subtilis* on surface of concrete could solve the problem of high cost in maintenance and at the same time, repair the concrete structure. *B. subtilis* fills the micro-crack by precipitating  $\text{CaCO}_3$  until it super-saturated in the void spaces. Although *B. subtilis* has proved its ability to heal a micro-crack on the

concrete surface by precipitating the  $\text{CaCO}_3$ , this application is merely for micro-crack remediation as a post-treatment method after the cracking occurred (Muynck et al., 2008; Raijiwala, 2008). Hence, a study on the efficiency of these biotic and abiotic factors is needed, since, these factors are among important roles for the survival and the growth of *B. subtilis* in order to enhance the micro-crack remediation treatment (Bhaskar, 2016).

In this study, the most ideal condition for each biotic and abiotic factor to enhance the efficiency of *B. subtilis* to treat WCA was investigated. This study was done to avoid internal formation of micro-crack due to the DEF in recycled cement mortar prior to its application as a part of the raw materials.

### **1.3 Objectives of Study**

The research objectives are as follows:

- a) To determine the characteristics of WCA from CDW.
- b) To determine the efficiency of WCA treatment by *B. subtilis* on the effect of *B. subtilis* concentration, *B. subtilis* : WCA ratio, zeta potential and retention time.
- c) To evaluate the compressive strength of recycled cement mortar (RCM) produced from the treated WCA (TWCA).

### **1.4 Significance of Study**

Directly utilised WCA as a cement mortar material without any treatment might provide a conducive environment for DEF to develop internally with high sulphate and water absorption due to the high attached cement paste in WCA. The

application of biological treatment on WCA could potentially treat the attached cement paste. In this study, utilisation of *B. subtilis* on WCA potentially reduces the sulphate content prior to the massive recrystallisation of DEF in the ITZ and enhanced physical characteristic (water absorption and density). This, treated TWCA would further prevent the problem of micro-crack formation due to the reduction of these factors and simultaneously prolongs the life span of RCM.

Furthermore, this pre-treatment product could be applied along with Industrialised Building System (IBS) approach since the concrete would be manufactured off-site before it is deposited to the construction site. Thus, by introducing the recycling approach on waste concrete, it is not only reducing the amount of CDW dumped at the landfills or along the roadsides, it also saves the environment by reducing the natural resource dependency and energy consumption, as well as overcoming the drawback characteristics of WCA.

## **1.5 Scope of Study**

This study focused on WCA generated from CDW to produce a newly product, namely RCM. The research intended to identify the characteristics of physical, mineralogical, chemical, soluble sulphate content and leaching behaviour of inorganic elements as well as a total amount of attached cement paste consisted in WCA. To identify its characteristics, the crushed waste concrete was sieved into a fine aggregate (WCA) with sizes ranging from 0.3 mm to 5 mm prior to the particle size distribution analysis.

The efficiency of *B.subtilis* for WCA treatment was determined based on *B. subtilis* concentration, *B. subtilis*: WCA ratios, zeta potential, retention times, filtrate quality and physical characteristic. The quality of the filtrate was evaluated in order to analyse any harmful elements leached out from the treatment. The TWCA was subsequently utilised as a cement mortar material for the production of RCM, replacing the sand typically used in concrete production. The compressive strength of the RCM was also evaluated as a comparison with a normal cement mortar (control).

## **1.6 Novelty and Gaps of Study**

The drawbacks characteristics of WCA on density, water absorption and sulphate content have restrained to be recycled which influenced by the attached cement paste on the surface of WCA (Evangelista & de-Brito, 2007; Juan & Gutierrez, 2009, de-Brito & Nabajyoti, 2013). These characteristics could provide a conducive environment for the formation of DEF in ITZ. Massive distribution of DEF caused propagation of micro-crack and subsequently expanded the crystal up to the concrete and mortar surface (Shi et al., 2012). Natural process in cement- based material (concrete and mortar) results in release of sulphate and subsequently enhanced the DEF (Lee et al., 2005; Graf, 2007). Thus, recrystallization of DEF increases the crystal volume in void spaces, which finally triggers the swell pressure and causing mass loss and consequence of reducing the material strength (Hu et al., 2006).

The use of *B. subtilis* on surface of concrete has been introduced and investigated by many researchers to solve the micro-crack formation and repair the concrete structure. *B. subtilis* precipitating  $\text{CaCO}_3$  and fully packed present of micro-crack in WCA. This application could improve density and material strength but is

merely for micro-crack remediation as post- treatment method after the cracking occurred (Raijiwala, 2008).

Besides precipitating  $\text{CaCO}_3$ , *B. subtilis* could consume the sulphate for its biochemical reaction and metabolic activities (Ghosh et al., 2005). Therefore, a study on the efficiency of biotic and abiotic factors is needed, since these factors are among important roles for the growth of *B. subtilis* in micro-crack (Bhaskar, 2016). While additional procedure of pre-saturated and air-dried in WCA treatment, could improve water absorption value of WCA (Angulo et al., 2012). Hence, combination of this procedure with utilisation of *B. subtilis* could improve the drawback characteristics before the recycle WCA can be used.

The treated WCA would further prevent the problem of micro-crack formation due to the reduction of sulphate and simultaneously prolongs the life span of RCM. Recycled WCA not only reducing the amount of CDW dumped at the landfills or along the roadsides, it also saves the environment by reducing the natural resource dependency and energy consumption, as well as overcoming the drawback characteristics of WCA.

## **CHAPTER 2**

### **LITERATURE REVIEW**

#### **2.1 Construction and Demolition Waste (CDW)**

Construction and demolition waste (CDW) is a solid waste generated from any construction and demolition activities due to the improvement, preparatory, repair or alternation work (ACT 627, 2007). These activities included civil and building construction, renovation and demolition (site clearance, roadwork, land excavation and formation) (Shen et al., 2004). It is also categorised as an inert solid waste and a complete loss of physical waste from construction sites (Kofoworola & Gheewal, 2009; Katz & Baum, 2011; Nagapan et al., 2012). Nowadays, construction industry plays a vital role in economic growth to improve quality of life by providing necessary infrastructures such as road, housing, commercial development and other facilities. In Malaysia, government has allocated 5, 555 projects for development sector by improving the residential, non-residential building projects, social amenities and infrastructure projects (CIDB, 2012). Due to the circumstances, high demand for construction development has generated high volume of CDW.

European statistic reported a total of 859 million tonnes of CDW which covered 32.9% of the total solid waste (Eurostat, 2011). Many countries recorded CDW have high number of annual generation (Table 2.1). In UK and US were 70 million tonnes and 323 million tonnes (EPA, 2004; Designing Building Wiki, 2017), respectively. Hong Kong Environment Protection Department reported municipal solid waste landfill had received 3, 158 tonnes/day of CDW (Hong Kong Waste Reduction, 2015). Meanwhile, Malaysia recorded a total of 100, 000 to 175, 000 tonnes

of waste and the amount is believed to be increasing each year (The Star Online, 2006). This waste was generated from CDW and is relatively high due to the contribution of construction project (Forsberg & Soukkoriipi, 2007). Azis et al. (2012) stated that waste material from CDW was 9 % by weight of purchase materials. The generation of CDW usually due to the problem on planning, material, human and environment such as frequently change on design (Zhao & Chua, 2003; Yupeng, 2011), low quality of materials (Nazech et al., 2008; Jameel et al., 2011), worker mistake (Polat & Ballard, 2004; Llatas, 2011), weak ability on site management (Zhao & Chua, 2003; Lu et al., 2011;) and bad weather (Senaratne & Wijesiri, 2008; Wahab & Lawal, 2011).

Table 2.1 Annual generation of CDW in four countries throughout the world

| Year | Amount             | Country  |
|------|--------------------|----------|
| 2000 | 70 million tonnes  | UK       |
| 2003 | 323 million tonnes | US       |
| 2007 | 1 million tonnes   | China    |
| 2012 | 4 million tonnes   | Malaysia |

### 2.1.1 Waste Disposal

High project development on housing, infrastructures or commercial buildings has generated high volume of CDW (Begum et al., 2006). In Malaysia, Bercham Landfill (Ipoh) and Jelutong Landfill (Penang) are legal dumping site collecting CDW (Mahayuddin et al., 2008; Nagapan et al., 2012). As reported in Malaysia Solid Waste and Public Cleansing Management Act 2007, CDW has considered as a controlled solid waste that needs efficiently dispose (Act 627, 2007). However, illegal dumping is considered as a common disposal practice in Malaysia (Yahaya & Narsen, 2008). Approximately, 42% from 46 illegal dumping sites in Johor district is CDW (Rahmat & Ibrahim, 2007). CDW was also illegally dumped along the roadside of Seberang



Perai, Pulau Pinang (Faridah et al., 2004). Murali (2011) reported that 30 tonnes of CDW was illegally dumped in tropical mangrove swamp near Bandar Hilir, Malacca (Appendix 1). While in 2012, CDW has illegally dumped near the roadside at Section 17, Petaling Jaya, Selangor (Appendix 2) (Tan, 2012).

Illegal dumping has governed public concerns owing to the risk on environment and human health (Faridah et al., 2004; Begum et al., 2006; Rahmat & Ibrahim, 2007). High transportation cost and distance of location project are among the reasons to select illegal dumping as a disposal practice. Illegal dumping occurs when the distance of project location was far from the gazetted landfill and the contractors refuse to expense additional cost especially for the waste management (Kamarul & Zuhairi, 2007). According to the Lestari Universiti Kebangsaan Malaysia (UKM) research, only 68 % of the contractors agreed to pay for services on collection and disposal (Pereira et al., 2005). Thus, providing a systematic and efficient waste management service such as re-using or re-cycling construction waste materials are required to minimise the amount of CDW being dumped at illegal sites or landfills.

### **2.1.2 Waste Concrete as Major CDW**

There are many types of physical waste generated from CDW e.g. concrete, ceramic, metal, plastic, brick, tile, asphalt, gypsum, paper, glass, sand and wood (Jang & Townsend, 2001; Faridah et al., 2004; Bianchini et al., 2005; Oikonomou, 2005; Vegas et al., 2011; Lachimpadi et al., 2012; Lee et al., 2013). Based on previous findings, it was found that concrete dominated the physical waste generated from CDW (Jang & Townsend, 2001; Faridah et al., 2004; Oikonomou, 2005; Vegas et al., 2011; Lachimpadi et al., 2012).

Moreover, Construction Industry Development Board (CIDB) has claimed concrete debris as major component generated from construction industry (Kamarul & Zuhairi, 2007). Therefore, introduction of Industrial Building System (IBS) by CIDB as an alternatives to re-enforce the commitment of construction industry towards sustainable development, in order to implement zero waste concept in construction industry.

### **2.1.3 Characteristics of concrete**

Concrete was generally used as a construction material in structural applications (Silva et al., 2014). Concrete is considered as the main waste generated among CDW due to the high demand in major infrastructure project, housing and commercial development (Belgium et al., 2010). Cement, sand and coarse aggregate are main material for production of concrete. Therefore, crushed waste concrete aggregate potentially have vary portion of cement attached with sand and/or coarse aggregate. Hence, the variation amount of the attached cement paste require to be analysis before conducting a treatment on crushed waste concrete which influence deterioration degree of concrete (Martin-Morales et al., 2011).

## **2.2 Recycling CDW as a Recycled Concrete Aggregate**

Many researchers attempted to create various types of CDW as recycled material as it is applicable to numerous functions such as pedestrian blocks (Kamarul & Zuhairi, 2007), unbound structure layer of road (Vegas et al., 2011), mortar (Bianchini et al., 2005) and concrete (Katz, 2003). Besides waste concrete, there are many types of CDW potentially to be recycled, for instance, bricks (Poon & Chan, 2006; Umami et al., 2008; Bektas et al., 2009), ceramic (Binici, 2007; Corinaldesi et

al., 2010), plastic (Batayneh et al., 2007; Ismail & Al-Hashmi, 2008; Choi et al., 2009; Tan & Du, 2013) and glasses (Hui & Sun, 2011; Matos & Sousa-Coutinho, 2012; Tan & Du, 2013).

However, most of the above-mentioned works focussed on examining suitable replacement materials for concrete production, either a cement or a concrete aggregate. Yet, the production of the cement deals with few challenges, particularly on CO<sub>2</sub> emission and somewhat costly in energy supply (Schneider et al., 2011). Owing to those factors, many studies have been directed to investigate various types of CDW in order to produce high performance recycled product which simultaneously reduce the dependency on cement as a raw material. The replacement materials for cement portion require a pozzolana component as a cementitious feature, which acts as a binder.

Replacement materials such as wood ash, plastic, glass, ceramic and brick have similarity on certain physical and chemical features of cement and concrete aggregate. These replacement material has fine particle size, low water absorption, low unit weight and specific gravity (Cheah & Ramli, 2012). Besides, rich carbon, silica, alumina, portlandite and lime, which relative to neat cement (Udoeyo et al., 2006). However, several factors have limited the capability of these replacement materials to be recycled, although the presence of pozzolana component (Table 2.2).

Table 2.2 Limitations of different types of CDW for cement and concrete aggregate replacement

| <b>Replacement Material</b> | <b>Limitation</b>   | <b>References</b>  |
|-----------------------------|---|--|
| Marble powder               | Irregular shape, fineness particle size ( $< 7\mu\text{m}$ ) resulted demanded water for cement hydration   | Corinaldesi et al. (2010)<br>Cheah & Ramli (2012)  |
| Wood Ash                    | High combustible organic content caused water demand for cement hydration<br>Prolong setting time due difficulty to dilution for cement hydration<br>Late pozzolanic reaction caused retardation cement hydration<br>Act as filler not binder material<br>Increased replacement material percent reduce compressive strength  | Udoeyo et al. (2006)<br><br>Abdullahi (2006)<br>Elinwa & Ejeh (2004)<br>Udoeyo et al. (2006)<br>Cheah and Ramli (2012)   |
| Plastic                     | Hydrophobic, sharper edge, angular size, slippery surface texture caused difficulty for cement hydration<br>High total surface area caused high water absorption<br>Short fibres acted as bridge-action of crack cause post-cracking on specimen<br>Low adhesive strength<br>Increased replacement portion reduced compressive strength<br>Low adhesive strength results low compressive strength | Ismail & Al-Hashmi (2008); Choi et al. (2009)<br>Albano et al. (2009)<br>Lucolano et al. (2013)<br>Ismail and Al-Hashmi (2008)<br>Batayneh et al. (2007)<br>Saikia & De-Brito (2012) |
| Glass                       | ASR reaction between high silicate of glass and high alkali of aggregate<br>Irregularity surface, smooth and sharper edge of particle<br>Low adhesive strength<br>Micro-crack during crushing process lead ASR expansion  | Bazent et al. (2000);<br>Hui & Sun (2011)<br>Batayneh et al. (2007);<br>Tan & Du (2013)  |
| Ceramic                     | Required huge amount of super-plasticizer   | Binici (2007)  |
| Brick                       | High water absorption due to high porosity<br>Angular shape caused difficulty for cement hydration<br>Compressive strength not affected by percent of replacement ratio   | Bektas et al. (2009)   |

These limiting factors have caused high water absorption, crack formation, expansion of alkali silicate reaction (ASR) and low adhesive strength between replacement materials, cement and /or aggregate. These conditions led to air entraining action and water infiltration in void spaces, which eventually promoting an extensive ASR and micro-crack formation. Furthermore, these replacement materials depend on a super-plasticiser to overcome the drawback properties where it could enhance the adhesive strength at Interfacial Transition Zone (ITZ), producing high compressive strength of recycled products (Shayan, 2002; Corinaldesi et al., 2010; Cheah & Ramli, 2012).

Although these materials are potentially useful as replacements of cement or concrete aggregate, these limiting factors have restrained the value-added of these materials on the production of recycled concrete and mortar. Several works have reported the effect of using these materials on a long-term durability, compressive strength and toxic chemicals they may possess (Batayneh et al., 2007; Binici, 2007; Ismail & Al-Hashmi, 2008; Bektas et al., 2009; Corinaldesi et al., 2010; Frigione, 2010; Cheah & Ramli, 2011; Saikia & de-Brito, 2012; Wang & Mayer, 2012; Tan & Du, 2013).

IBS system has shown that the major CDW is waste concrete (Lachimpadi et al., 2012). Therefore, the use of waste concrete as a replacement material on concrete aggregate portion could reduce the amount of waste generation, of which a sustainable development in construction industry could further be achieved. Also, many studies have demonstrated that recycled waste concrete that has been used as a replacement material has no adverse effect on the mechanical strength of the recycled product when

approximately 30% of waste concrete was mixed with coarse aggregate (Batayneh et al., 2007; Evangelista & de-Brito, 2007).

For recycling purpose, a total construction budget has been saved up to 2.5 % when a recycling approach on CDW was introduced (Begum et al., 2007). Besides, the dependency on natural resources can be reduced and subsequently, the cost of raw materials also can be reduced. In addition, production of aggregate and cement at a quarry requires an acid washing to remove any unwanted element and a frequent washing surface run-off of silt, sand or gravel in drainage pipes may lead to the blockage. Thus, the production of aggregate and cement at the quarry does not only cause the water wastage but also could harm the biotic life due to the toxic chemical utilisation (Kin-sun, 2004). Moreover, excessive energy consumption (up to 40% of total energy needed) for the production and transportation of aggregate and cement is another reason as to why a recycling approach should be considered (Reddy & Jagadish, 2003; Zapata & Gambatese, 2005).

Construction industry has been widely utilised recycling approach. For example, approximately 30 million tonnes of recycled concrete aggregate were utilised in UK (Limbachiya et al., 2004). While, In USA, Federal Highway Administration has utilised CDW as a recycled concrete and pavement (Schimmoller et al., 2000). Recycled concrete aggregate also can be used for the production of lightweight building materials as well as high strength concrete (Limbachiya et al., 2000).

## **2.3 Characteristics and Effects of Using Waste Concrete Aggregate (WCA) from CDW**

Waste concrete aggregate (WCA) is a complete loss of physical solid waste removed from construction sites to disposal sites (Nagapan et al., 2012). Among the generation factors for such solid waste are carelessness, over-ordering and over-filling to container of lifting equipment while concrete casting (Foo et al., 2013). This poor management by unskilled work force causes a lower productivity and workmanship at construction sites (Sambasivan & Soon, 2007). Subsequently, WCA is highly generated compared to other types of CDW (Lachimpadi et al., 2012).

### **2.3.1 Physical Characteristics**

Density is one of the fundamental parameters and determines concrete mixes design which directly controls other concrete properties. It is also the most common method to characterize the aggregates. Low density of WCA is related to high content of attached cement paste (Martin-Morales et al., 2011). The reasons are the porosity and less density of attached cement paste adhered onto the surface of WCA which simultaneously lead to the low density of WCA (Poon et al., 2006; de-Brito & Saikia, 2013). Recycling procedure of WCA had discussed by Silva et al. (2014) that may affect the density of WCA. Crushing the concrete rubble into smaller sizes is the first step in recycling procedure.

The recycling procedure for waste concrete requires a pass through primary and secondary stages of crushing process (Limbachiya et al., 2007; Ferreira et al., 2011; Fonseca et al., 2011). A jaw crusher is typically used in the primary stage, while, a cone crusher and impact crusher are used during the secondary crushing stage to

produce rounder and less sharp particles of fine WCA. The number of crushing stages influences the density of WCA, of which more crushing stages will cause a lower density of WCA (Nagataki et al., 2000; Nagataki et al., 2004; Gokce et al., 2011). This scenario is due to the cumulative breaking up of adhered cement paste on the surface of the WCA. Therefore, Kasai (2004) has suggested that the crushing stages should not be too few or too many, otherwise, the produced fine WCA has low density due to the high amount of attached cement paste. Thus, the density is expected to be low due to the accumulation of attached cement paste in fine WCA.

Generally, low density WCA has an impact on mechanical strength (Etxeberria et al., 2007; Tam et al., 2008; Padmini et al., 2009). High amount of cement is required due to the low particles mass of WCA to occupy the void spaces of recycled product specimen (Martin-Morales et al., 2011). Previous researchers have demonstrated that mechanical strength of different concrete mixes decreased when saturated surface dry density decreased (Nagataki et al., 2004; Andreu & Miren, 2014). It can be concluded that the mechanical strength is clearly influenced by the density of aggregates. Meanwhile, Santos and colleagues (2002) highlighted the concrete with higher amount of attached cement paste (49.4%) tend to have lower mechanical strength i.e. 45 MPa compared to the concrete with lower cement paste (36.3%), of which the strength was 56 MPa. Thus, the amount of attached cement paste onto WCA influences its density which simultaneously affects the mechanical strength of the concrete (Nagataki et al., 2000; Otsuki et al., 2003; Shayan & Xu, 2003; Sanchez et al., 2004; Isgor & Razaqpur, 2005).



Water absorption is another parameter to be considered when using WCA as similar factors as those designated for density (Silva et al., 2014). The water absorption of an aggregate is directly related to its porosity which is highly dependent on total amount of attached cement paste (Khalaf & DeVenny, 2004; Poon et al., 2006; Etxeberria et al., 2007; Juan & Gutierrez, 2009). The above statement is also supported by de-Brito and Saikia (2013) where they stated that WCA has higher water absorption value than that of natural aggregate due to the presence of attached cement paste. As mentioned earlier, the more the crushing stages, the higher the amount of attached cement paste. Thus, the presence of attached cement paste with high porosity causes higher amount of water being absorbed by WCA.

High water absorption in WCA definitely influences the concrete mixing process. It causes more water to be consumed which affects the water/cement ratio and eventually gives impact to the workability and the hardened process of fresh concrete (Santos et al., 2002; Sanchez et al., 2004; Isgor & Razaqpur, 2005; Padmini et al., 2009). Furthermore, excessive water leads to the formation of a thin layer of cement slurry on the surface of WCA which permeates into porous old cement mortar, micro-crack and voids (Tam et al., 2007). Although cement slurry develops a strong ITZ between cement paste and WCA, but it enhances the deterioration of concrete following the formation of ettringite, portlandite and calcium silicate hydrate (C-S-H).

Attached cement paste onto WCA surface was technically impossible to be removed from the aggregate due to the cost and technical aspects (de-Brito & Saikia, 2013). Therefore, concrete mixes design requires additive stages which is pre-soaking, in order to prevent excessive suction of mixing water during the mixing process

(Zaharieva et al., 2003). Pre-saturated of WCA followed with 30 min air –dried has demonstrated better mechanical performance of recycled concrete as compared to the oven dry alone (Poon et al., 2004). Angulo and colleagues (2010) showed that soaking WCA with water has a great effect on water absorption value i.e. 17% reduction from the initial value. Therefore, pre-treatment with soaking and air-dried procedures could be improve water absorption of WCA.

### **2.3.2 Mineralogical and Chemical Characteristics**

Solid waste of CDW is usually dumped at disposal sites without any segregation process and it is important to determine the mineralogical characteristic of WCA due to the possible hazardous minerals (amianthus) present in asbestos. Moreover, the mineralogical characteristics of WCA influence the procedure of recycled concrete production such as water/cement (w/c) ratio and curing day, in order to achieve an equal performance as an ordinary concrete (Limbachiya et al., 2007). In laboratory, X-ray diffractometry (XRD) technique is commonly to detect types of minerals constituted in WCA.

Generally, minerals such as calcite, quartz, ettringite and portlandite are commonly detected in WCA (Bianchini et al., 2005; Sani et al., 2005; Vegas et al., 2011). Quartz present in natural rock, while, calcite is from the cement (Angulo et al., 2009). WCA tends to have low portlandite as compared to natural aggregate due to the light pozzolanic effect (Sani et al., 2005). Low portlandite in WCA causes an un-stable of ettringite and subsequently increases the solubility of sulphate ion in pore water occupied in void spaces of concrete (Vegas et al., 2011). Besides, high calcite content indicated carbonated WCA due to the rich CaO (Angulo et al., 2009)

Although density and water absorption are vital parameters that control the workability of concrete and mortar, inorganic element within WCA should also need to be concerned. Many researchers have pointed out that the CDW has feasibility threat to the environment and human health (Van der Sloot, 2000; Roussat et al., 2008; Michelis et al., 2009; Jimenez et al., 2012) . Nowadays, recycled concrete and mortar are utilised in road construction and building structures. However, during their service life span, environmental impact from rain, surface water and ground water are potentially in contact with existing inorganic elements from the WCA. The environmental impact is not rely on the total content of pollutants but depends on the amount of the pollutants able to be dissolved by water and subsequently leaches out from the medium (Kosson et al., 2002; Polo et al., 2005). This phenomenon finally pollutes the surface or ground water. Thus, potential inorganic elements leaching out from the waste concrete and solubility of each element are among crucial parts to assess environmental impact. Furthermore, EU commission had claimed that CDW is one of the main waste streams to be dumped in landfills (European Commisison Report, 2002)

Inorganic elements in WCA are examined by X-ray fluorescence (XRF) spectrometry. Since the environmental impact is correlated with ions that leached out from WCA, therefore, leachability of ions is definitely related to the chemical element of WCA (Limbachiya et al., 2007). Previous research on pH-dependent batch leaching test i.e. CEN/TS 14429 (2005) in order to determine the leaching behaviour of chemical elements from WCA. This method used  $10 \pm 0.3$  ml/mg as liquid to solid ratio for 48 hour agitation period with deionized water as the extraction medium

(Engelsen et al., 2009, 2010; Mulugeta et al., 2011; Galvin et al., 2012; Galvin et al., 2013; Solpuker et al., 2014).

XRF analysis and leaching test from previous studies have shown that calcium oxide (CaO), aluminium oxide (Al<sub>2</sub>O<sub>3</sub>) and silicon dioxide (SiO<sub>2</sub>) are the main chemical elements leached out from concrete aggregate (Limbachiya et al., 2007; Marshitah et al., 2009; Angulo et al., 2009). Few elements such as, SO<sub>4</sub><sup>2-</sup>, Cr<sub>2</sub>O<sub>3</sub> and SrO are only detected as trace elements (Bianchini et al., 2005; Limbachiya et al., 2007; Mulugeta et al., 2011). While, abundant with element of Al<sub>2</sub>O<sub>3</sub> in waste concrete due to the clay-based minerals, which is the main element of attached cement (Bianchini et al. (2005).

### **2.3.3 Leachability**

A serious matter needs to be considered concerning the use of WCA in construction industry is it may contain leachable pollutants (trace inorganic element) to human health and the environment (Barbudo et al., 2012). Leached trace element such as lead, copper, cadmium and zinc in urban runoff caused accumulated in buildings, particulates in vehicle brake and air pollution (Herngren et al., 2005; Boving et al., 2008). The presence of sulphate element in quartz has a cathartic effect in human health when its presence is in excessive amount (Donaldson et al., 2001). Leaching behaviour by testing the materials under pH-controlled conditions has considerably increased following the fact that pH is one of the major parameters regulating the release of elements from the solid phase into solution. The test can be used to identify any changes in leaching behaviour. The long-term potential risks are generally to the elements of Mo, V, As, Cr, B and Sb, which, the leachability of these elements is high

at mild alkaline to neutral pH. Furthermore, these elements are the most frequently found in cement-based products after long term exposure under environmental condition.

Most inorganic elements in WCA such as Ca, Zn and Cr are pH-dependent (Engelsen et al., 2009, 2010; Solpuker et al., 2014) with only Se shows no pH-dependency behaviour for it could be leached out from WCA (Galvin et al., 2012). Leaching trends for each element are different and depend on the solubility of minerals or by the solubility of metal hydroxide (Engelsen et al., 2009). For example, low pH of solution would lead to unstable stage of calcite and simultaneously causes high concentration of Ca in leachate (Solpuker et al., 2014). While higher pH of solution (more than 11) would cause dissolution of Fe-monosulphate (FeS) and amorphous iron(III) hydroxide ( $\text{Fe}(\text{OH})_3$ ) which further causing the concentration of Fe to incline (Solpuker et al., 2014).

#### **2.3.4 Soluble Sulphate Content**

In soil, sulphates present in the form of calcium, magnesium, sodium and potassium (Prasad et al., 2006). Ammonium sulphate, for example, is frequently found in agricultural soil and water. In WCA, sulphate is commonly present in attached cement paste and concrete aggregate clinker (Juan & Gutierrez, 2009). However, Scanning Electron Microscope (SEM) analysis has proved that sulphate was found in ettringite mineral as a part of attached cement paste components (Lee et al., 2008). Sulphate from aggregate clinker reacted spontaneously with anhydrous calcium aluminate from cement particularly for cement hydration (Pavoine et al., 2006; Tam et al., 2009). However, enhancement of sulphate content in concrete has caused micro-